

(19) World Intellectual Property
Organization
International Bureau



(43) International Publication Date
17 June 2004 (17.06.2004)

PCT

(10) International Publication Number
WO 2004/050045 A1

(51) International Patent Classification⁷:

A61K 7/00

(84) Designated States (*regional*): ARIPO patent (BW, GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, RO, SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

(21) International Application Number:

PCT/US2003/038455

(22) International Filing Date: 3 December 2003 (03.12.2003)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

60/431,141 4 December 2002 (04.12.2002) US

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(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.

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- *of inventorship (Rule 4.17(iv)) for US only*

Published:

- *with international search report*
- *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 2004/050045 A1

(54) Title: VESICLE AND MICROEMULSION COMPOSITIONS OF POLAR ORGANIC OILS BASED ON SILICONE SURFACTANTS

(57) Abstract: Vesicle and microemulsion compositions comprising a silicone surfactant, a co-surfactant, a polar organic oil, and water are disclosed. A method for forming vesicles from a microemulsion comprising a silicone surfactant, a co-surfactant, a polar organic oil, and water is also disclosed. The vesicle and microemulsion compositions are useful in a variety of personal care applications.

VESICLE AND MICROEMULSION COMPOSITIONS
OF POLAR ORGANIC OILS
BASED ON
SILICONE SURFACTANTS

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The present invention relates to vesicle and microemulsion compositions comprising a silicone surfactant, a co-surfactant, a polar organic oil, and water. The present invention further relates to a method for forming vesicles from a microemulsion comprising a silicone surfactant, a co-surfactant, a polar organic oil, and water.

10 Vesicles based on silicone surfactants were first disclosed in U.S. Patents 5,364,633 and 5,411,744 by Hill and Snow. The '633 and '744 teaches the preparation of vesicles in water formed solely from certain siloxane surfactants. The vesicles were able to entrap up to 10 weight % of either a water soluble or water insoluble substance, depending on the method of preparation of the vesicle compositions.

15 U.S. 5,958,433 discloses dispersions of a water-immiscible phase, in an external aqueous phase by means of lipid vesicles comprising at least one silicone surfactant. The '433 patent further discloses the use of an ionic amphiphilic lipid for addition to the silicone surfactant to inhibit flocculation of the dispersion.

20 U.S 5,919,487 teaches nanoparticles, and in particular nanocapsules, with a lamellar coating obtained from a silicone surfactant. The nanoparticles of the '487 patent range in size from 10 to 1000 nm and are composed of a polymer encapsulating an oily phase and coated with a lamellar coating comprising at least one silicone surfactant.

25 Silicone surfactants have also been used to prepare microemulsions of silicones, and in particular volatile silicones. For example, U.S. Patents 5,705,562 and 5707,613 to Hill teaches clear silicone microemulsions of volatile silicones that form spontaneously, i.e., without the need for high shearing techniques, based on certain silicone surfactants. U.S 5,623,017 to Hill further teaches clear silicone gels of volatile siloxanes based on silicone surfactants.

30 U.S. 6,120,778 teaches transparent nanoemulsions of various oils based on silicone surfactants. The '778 patent further teaches the addition of amphiphic lipids to its compositions.

Commonly assigned co-pending U.S application 09/ 917,322 discloses microemulsions of volatile silicones stabilized by silicone surfactants, and optionally comprising certain co-surfactants.

While the use of silicone surfactants to prepare vesicles and microemulsions represent significant advances in the art, there is a need for additional improvements. In particular, vesicles formed from silicone surfactants can be limited in the amount of substance that can be entrapped in the lamellar phase of the vesicle. The benefits for using vesicles in various personal care formulations to deliver active materials, such as moisturizers, vitamins are well established. The efficiency of such formulations could be further enhanced if the concentration of the entrapped substance in the lamellar phase of a silicone surfactant based vesicle could be increased. In particular, there is a need to entrap high loadings of polar organic oils within the lamellar phase of silicone surfactant based vesicles.

The present inventors have discovered the addition of certain co-surfactants enhances the ability of vesicles based on silicone surfactants to entrap higher loadings of polar organic oils. The present inventors have further discovered the combination of certain co-surfactants and silicone surfactants further provides microemulsion compositions of polar organic oils, based on the selected concentration of the silicone surfactant/co-surfactant, polar organic oil and water. The inventive microemulsion compositions, upon dilution with water, produce vesicle-based compositions wherein the polar oil is entrapped with the lamellar phase of the vesicle.

The present invention provides compositions comprising a silicone surfactant, a co-surfactant, a polar organic oil, and water. In one embodiment, certain concentration ranges of the silicone surfactant, co-surfactant, polar organic oil, and water provide lamellar phase liquid crystalline structured compositions. In another embodiment, certain concentration ranges of the silicone surfactant, co-surfactant, polar organic oil, and water provide microemulsion compositions.

The present invention further provides a method for forming vesicles containing a silicone surfactant, co-surfactant, polar organic oil, and water comprising first forming a microemulsion of the silicone surfactant, co-surfactant, polar oil, and water, and then diluting the microemulsion with additional water to form vesicles.

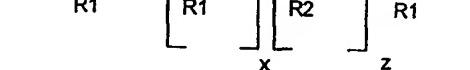
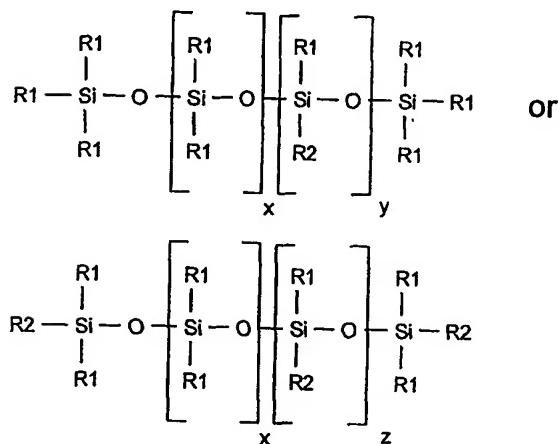
The present invention relates to compositions comprising;

- (A) a silicone surfactant,
- (B) a co-surfactant,
- (C) a polar organic oil, and
- (D) water.

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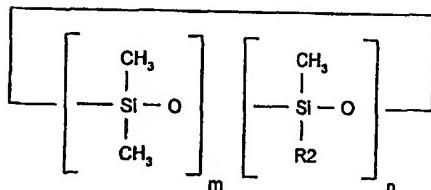
In one embodiment, certain concentration ranges, (discussed supra) of the silicone surfactant, co-surfactant, polar organic oil, and water provide lamellar phase liquid crystalline structured compositions. In another embodiment, certain concentration ranges (discussed supra) of the silicone surfactant, co-surfactant, polar organic oil, and water provide microemulsion
10 compositions.

Component (A) is a long chain or high molecular weight silicone polyether and can have a structure represented by:



A cyclic polyether of the type shown below can also be used.

15

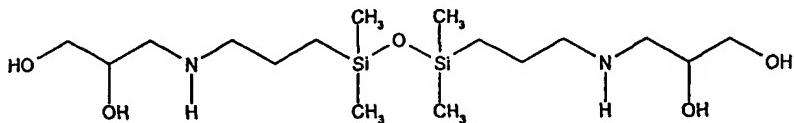


In these structures, R1 represents an alkyl group containing 1-6 carbon atoms such as methyl, ethyl, propyl, butyl, pentyl, and hexyl; R2 represents the group - $(CH_2)_aO(C_2H_4O)_b(C_3H_6O)_cR3$; x has a value of 1-1,000, alternatively 1 - 500, or

alternatively 10 - 250; y has a value of 1-500, alternatively 1- 100, or alternatively 2 - 50; z has a value of 1-500, or alternatively 1 - 100; m has a value of 3-5; n is one; a has a value of 3-6; b has a value of 4-20; c has a value of 0-5; and R3 is hydrogen, a methyl group, or an acyl group such as acetyl. Typically, R1 is methyl; b is 6-12; c is zero; and R3 is hydrogen.

5 The silicone surfactants can be prepared by any of the techniques known in the art, and many are commercially available. Representative commercial silicone surfactants suitable in the present invention include DC 5329 and DC 5211 (Dow Corning Corporation, Midland, MI)

Component (B) is a co-surfactant selected from the group of a monohydroxy
10 alcohol, an organic diol, an organic triol, an organic tetraol, a silicone diol, a silicone triol, a
silicone tetraol, and a nonionic organic surfactant. Some representative examples of the
cosurfactant component (B) include monohydroxy alcohols such as methanol, ethanol, and 2-
propanol; organic diols such as ethylene glycol, propylene glycol, and 1, 2 hexanediol;
organic triols such as glycerol; organic tetraols such as pentaerythritol and 1,2,3,6-hexane
15 tetraol; and a silicone tetraol such as shown below.



The nonionic surfactant should be a non-silicon atom containing nonionic emulsifier.
Typically the nonionic surfactant are alcohol ethoxylates R4-(OCH₂CH₂)_dOH, and in
20 particular are fatty alcohol ethoxylates. Fatty alcohol ethoxylates typically contain the
characteristic group -(OCH₂CH₂)_dOH which is attached to fatty hydrocarbon residue R4
which contains eight to twenty carbon atoms, such as lauryl (C₁₂), cetyl (C₁₆) and stearyl
(C₁₈). While the value of "d" may range from 1 to about 100, its value is typically in the
range of 2 to 40. Some examples of suitable nonionic surfactants are polyoxyethylene (4)
25 lauryl ether, polyoxyethylene (5) lauryl ether, polyoxyethylene (23) lauryl ether,
polyoxyethylene (2) cetyl ether, polyoxyethylene (10) cetyl ether, polyoxyethylene (20) cetyl
ether, polyoxyethylene (2) stearyl ether, polyoxyethylene (10) stearyl ether, polyoxyethylene
(20) stearyl ether, polyoxyethylene (21) stearyl ether, polyoxyethylene (100) stearyl ether,
polyoxyethylene (2) oleyl ether, and polyoxyethylene (10) oleyl ether. These and other fatty

alcohol ethoxylates are commercially available under names such as ALFONIC®, ARLACEL, BRIJ, GENAPOL®, LUTENSOL, NEODOL®, RENEX, SOFTANOL, SURFONIC®, TERGITOL®, TRYCOL, and VOLPO.

The co-surfactant can also be selected from cationic organic surfactants.

5 Representative cationic surfactants suitable for use as a co-surfactant in the present invention are described in U.S. 6,071,975, which is hereby incorporated by reference. Preferable cationic surfactants are selected from quaternary ammonium salts containing at least one organic group having at least 6 carbons, or alternatively containing a organic group derived from a fatty alcohol, such as dodecyltrimethylammonium bromide

10 The co-surfactant can also be selected from anionic organic surfactants.

Representative anionic organic surfactants suitable for use as a co-surfactant in the present invention are described in U.S. 6,071,975, which is hereby incorporated by reference.

Alternatively, the co-surfactant is an organic diol having the general formula R4-CH₂(OH)CH₂OH, where R4 is an alkyl hydrocarbon group containing 2 – 20 carbons,

15 alternatively 2 – 12 carbons, or alternatively 2 – 6 carbons. A preferred organic diol is 1-2 hexanediol.

The present invention does not encompass amphiphilic surfactants, as disclosed in U.S. Patents 5,958,433 and 6,120,778 as a possible selection of co-surfactant (B).

Component (C) is a polar organic oil. As used herein, a polar organic oil is any oil, or combination of oils. Typically, the polar organic oil is derived from an animal, a vegetable, or a mineral source. Modern cosmetic oils are most representative of the polar organic oil, and among the more common organic oils known to be safe for cosmetic purposes are almond oil, apricot kernel oil, avocado oil, cacao butter (theobroma oil), carrot seed oil, castor oil, citrus seed oil, coconut oil, corn oil, cottonseed oil, cucumber oil, egg oil, jojoba oil, lanolin oil, linseed oil, mineral oil, mink oil, olive oil, palm kernel oil, peach kernel oil, peanut oil, rapeseed oil, safflower oil, sesame oil, shark liver oil, soybean oil, sunflower seed oil, sweet almond oil, tallow (beef) oil, tallow (mutton) oil, turtle oil, vegetable oil, whale oil, and wheat germ oil.

Polar organic oils, as used herein, does not encompass silicone oils, that is oils based on polyorganosiloxanes.

The common assignee's U.S. Patent 5,948,855 (September 7, 1999), also contains an extensive list of some appropriate oil soluble active ingredients such as vitamins and drugs

which can be used in the oil phase of the water-in-oil emulsion, among which are vitamins, including but not limited to, Vitamin A₁, RETINOL, C₂-C₁₈ esters of RETINOL, Vitamin E, TOCOPHEROL, esters of Vitamin E, and mixtures thereof. RETINOL includes trans-RETINOL, 13-cis-RETINOL, 11-cis-RETINOL, 9-cis-RETINOL, and 3,4-didehydro-

- 5 RETINOL. Other vitamins which are appropriate include RETINYL ACETATE, RETINYL PALMITATE, RETINYL PROPIONATE, α -TOCOPHEROL, TOCOPHERSOLAN, TOCOPHERYL ACETATE, TOCOPHERYL LINOLEATE, TOCOPHERYL NICOTINATE, and TOCOPHERYL SUCCINATE.

Preferably, the polar oil is vitamin E or an ester of vitamin E.

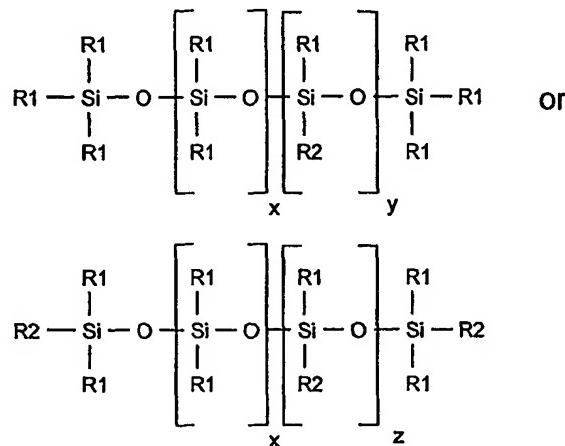
- 10 Components (A), (B), (C) and (D) can be combined at various weight percentages to produce compositions that are either i) vesicles having a lamellar phase structure or, ii) clear microemulsions. As used herein, the phrase "lamellar phase" describes the nature of the vesicle composition and is the accepted meaning in the art. The phrase "clear microemulsion" describes a emulsion composition containing particles that are smaller than 15 the wavelength of light. The microemulsion compositions of the present invention are be further defined by the definitions and requirements set forth in U.S. 5,705,562, which is hereby incorporated by reference.

- 20 The selection of the various weight percentages to produce compositions that are either i) vesicles having a lamellar phase structure or, ii) clear microemulsions can be done using a ternary phase diagram. Using this method, compositions are prepared by simply combining components (A), (B), (C) and (D) at various weight percentages. Excessive use of shearing techniques, or complicated mixing techniques are not necessary. Typically, the various components described infra are combined in a single container, shaken, or alternatively mixed with common lab mixers. The resulting composition is then 25 characterized as being either a single-phase system, i.e. a clear microemulsion, or a two-phase system, i.e. vesicles dispersed in water in which the vesicles have a lamellar phase structure. The formation of vesicles can be confirmed by techniques common in the state of the art. Typically, vesicles having a lamellar phase structure exhibit birefringence when examined with a cross polarizing microscope. For each composition prepared, a data point is 30 recorded on the ternary phase diagram designating that composition as being either as single-phase system or two-phase system. A representative example of a ternary phase diagram is shown in figure 1. In figure 1, compositions producing one- phase systems, i.e.

microemulsions, were designated by a spherical dot mark, whereas the compositions producing a two-phase system were designated by a triangular mark. Typically, a boundary line is found that separates the one phase and two-phase systems. Thus, it is typical to investigate many compositions that lie on or near the boundary of the phases to establish each 5 region in the ternary phase diagram for any given combination of components.

The first embodiment of the present invention provides vesicle compositions comprising;

A) 0.01 to 40 wt % of a silicone surfactant having a formula



10 where R1 represents an alkyl group containing 1-6 carbon atoms;
R2 represents the group $-(CH_2)_aO(C_2H_4O)_b(C_3H_6O)_cR_3$;
x is 1-1,000; y is 1-500; z is 1-500; a is 3-6; b is 4-20; c is 0-5;
and R3 is hydrogen, a methyl group, or an acyl group;

15 B) 0.01 to 60 wt % of a co-surfactant selected from the group of a monohydroxy alcohol, an organic diol, an organic triol, an organic tetraol, a silicone diol, a silicone triol, a silicone tetraol, a cationic organic surfactant, an anionic organic surfactant, and an nonionic organic surfactant;

20 C) 0.01 to 40 wt % of a polar organic oil

D) and water, in an amount to provide the balance of the composition to 100 % on a

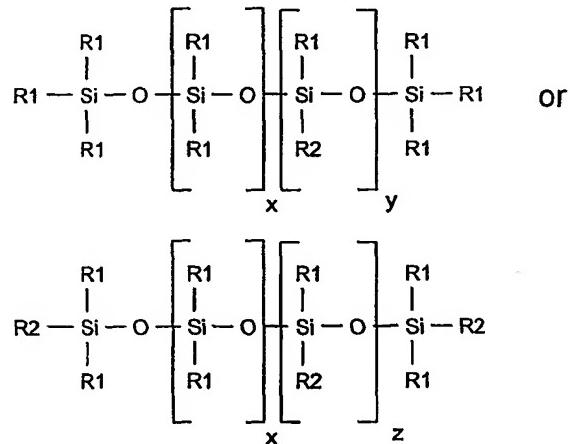
weight basis,

wherein the vesicle composition has a lamellar phase liquid crystalline structure.

5 Typically, the polar oil is entrapped in the lamellar phase liquid crystalline structure of the vesicle. The vesicle composition can contain as much as 20 weight %, alternatively 40 weight %, or alternatively 50 weight % of the polar organic oil wherein the polar organic oil is entrapped in the lamellar phase structure of the vesicle.

10 The second embodiment of the present invention provides microemulsion compositions comprising;

A) 7 to 90 wt % of a silicone surfactant having a formula



15 where R1 represents an alkyl group containing 1-6 carbon atoms;

R2 represents the group $-(\text{CH}_2)_a\text{O}(\text{C}_2\text{H}_4\text{O})_b(\text{C}_3\text{H}_6\text{O})_c\text{R3}$;

x is 1-1,000; y is 1-500; z is 1-500; a is 3-6; b is 4-20; c is 0-5;

and R3 is hydrogen, a methyl group, or an acyl group;

20 B) 0.01 to 70 wt % of a co-surfactant selected from the group of a monohydroxy

alcohol, an organic diol, an organic triol, an organic tetraol, a silicone diol, a silicone triol, a silicone tetraol, an cationic organic surfactant, an anionic organic surfactant, and an nonionic organic surfactant;

5 C) 0.01 to 60 wt % of a polar organic oil

D) and water, in an amount to provide the balance of the composition to 100 % on a weight basis,

10 wherein the composition is a clear microemulsion composition.

In either embodiment described infra, the weight ratio of components (A) to (B) can vary from 30:70 to 70:30, alternatively from 40:60 to 60:40, or alternatively from 45:55 to 55:45.

Figure 1 shows a ternary phase diagram for compositions formed from the
15 combinations of a 50/50 weight/weight mixture of a silicone surfactant (DC 5329, Dow Corning Corporation, Midland, MI) and a co-surfactant (1, 2 hexanediol), a polar organic oil (vitamin E), and water. This ternary phase diagram is a representative example of compositions of the present invention that can be formed from components (A) – (D) as defined infra. The region labeled as a one-phase system define the concentrations of
20 components (A) – (D) that produced microemulsion compositions, whereas the region labeled as a two phase system define the concentrations of components (A) – (D) that produced vesicles having the vitamin E entrapped with the lamellar phase structure of the vesicle.

Other organic polar oils can be selected for component (C), for example substituted or used in place of vitamin E from the example described in figure 1. Each organic polar oil
25 selected will produce a unique ternary phase diagram, given any combination of components (A), (B) and (D).

Alternatively, other co-surfactants can be selected, as defined infra, and used in place of the 1,2 hexanediol as used in the example for figure 1. As an additional example, figure 2 shows the ternary phase diagram for the combinations of a 50/50 weight/weight mixture of a
30 silicone surfactant (DC 5329, Dow Corning Corporation, Midland, MI) and a co-surfactant, ($C_6H_{13}(CH_2CH_2O)_2$, designated as C6E2 in figure 2 , a polar organic oil (vitamin E), and water.

One skilled in the art would be able to select the concentrations of components (A), (B) and (D) for any given polar organic oil (C), given the teaching of the present invention.

The present invention further provides a method for forming vesicles containing a silicone surfactant, co-surfactant, polar organic oil, and water comprising first forming a 5 microemulsion of (A) a silicone surfactant, (B) a co-surfactant, (C) an organic polar oil, and (D) water, and then diluting the microemulsion with additional water to form vesicles.

Components (A) – (D) are the same as defined infra. The microemulsion formed from components (A) – (D) is the same as defined above, and designated as a one-phase system in the ternary phase diagram. The formed microemulsion is then diluted with additional water 10 to form vesicles (as defined infra) wherein the polar organic oil is entrapped in the lamellar phase of the vesicles.

The microemulsions and vesicles prepared according to the invention can be used in various over-the-counter (OTC) personal care products. Thus, they can be used in antiperspirants, deodorants, skin creams, skin care lotions, moisturizers, facial treatments 15 such as acne or wrinkle removers, personal and facial cleansers, bath oils, perfumes, colognes, sachets, sunscreens, pre-shave and after-shave lotions, liquid soaps, shaving soaps, shaving lathers, hair shampoos, hair conditioners, hair sprays, mousses, permanents, depilatories, cuticle coats, make-ups, color cosmetics, foundations, blushes, lipsticks, lip balms, eyeliners, mascaras, oil removers, color cosmetic removers, and powders. The 20 microemulsion compositions are also useful as carriers for pharmaceuticals, biocides, herbicides, pesticides, and to incorporate water and water-soluble substances into hydrophobic systems.

Furthermore, it is anticipated that the vesicle and microemulsion compositions of the present invention can be combined with various other components to prepare the personal 25 care products described infra. These components include additional surfactants, moisturizers, pigments, sunscreens, fragrances, emollients, commonly used to formulate such personal care products.

EXAMPLES

Example I

5 A silicone polyether (DC 5329, Dow Coming Corporation, Midland, MI) 1 part, and 99 parts de-ionized water were added to a vial and hand-shaken for approximately 30 seconds. Particle size data revealed peaks at 1.072 μm and 0.1717 μm were observed. This shows bi-modal distribution of particle size.

10 Example II

5 parts DTAB (dodecyltrimethylammonium bromide) and 95 parts de-ionized water were added to a vial and hand-mixed until the DTAB was dissolved. 1 part DC 5329 was then added, and the vial was hand-shaken for 2 minutes. Particle size analysis revealed a single peak at 0.1085 μm . Compared to Example I, the larger particle distribution (peaked around 0.1717 μm) was eliminated. This shows the effect of addition of DTAB.

Example III

6 parts 1,2-hexanediol and 94 parts de-ionized water were added to a vial and stirred overnight on a stir plate to prepare a 6% 1,2-hexanediol aqueous dispersion. 1 part DC 5329 20 and 99 parts of the 6% 1,2-hexanediol aqueous dispersion were added to a vial and hand-shaken for approximately two minutes. Particle size data revealed a peak at 0.156 μm . This resulted in single particle size distribution and smaller particle size (compared to Example I).

Example IV

25 1 part vitamin E acetate and 4 parts DC 5329 were added to a vial, and this was rotated overnight. Using the 6% 1,2-hexanediol aqueous dispersion prepared in Example III, 1 part of the 20% vitamin E acetate in DC 5329 solution was combined with 99 parts 6% 1,2-hexanediol in water in a vial. This sample was rotated overnight, and particle size data revealed peaks at 2.761 μm and 0.381 μm . This resulted in smaller particle size distribution 30 (compare to Example V).

Example V (comparative)

Using the 20% vitamin E acetate in DC 5329 solution prepared in Example IV, 1 part of this solution and 99 parts de-ionized water were added to a vial. The vial was rotated overnight, and particle size analysis on the following day revealed peaks at 4.427 μm and

5 1.024 μm .

Example VI (comparative)

30 parts of vitamin E and 70 parts DC 5329 were added in a beaker to make a 30% vitamin E in 5329 solution. 10 parts of the 30% vitamin E in DC 5329 and 90 parts of di-ionized water were added to a vial. Handshaking produced a white precipitate at the bottom of the vial.

Example VII

10 parts of the 30% vitamin E in DC 5329 made in Example VI, 84% of de-ionized water, and 6% 1,2-hexanediol were added in a vial and hand shaken to form a uniform turbid dispersion. This shows the effect of 1,2-hexanediol on incorporating high concentration of vitamin E and the process of making.

Example VIII

20 *Preparation of Microemulsion based on Silicone Surfactants*

A 50/50 mixture of DC 5329 and 1,2-hexanediol was made by heating the DC 5329 for 45 seconds in a microwave oven and then adding the 1,2-hexanediol. The mixture was shaken and spun on a rotary wheel (Roto-Torque, Model # 7637-01) for thirty minutes. The 50/50 mixture was used at room temperature.

25 For each sample, a triangular graph was used to determine the desired percentages of each of the three components to be used. Using an analytical balance, samples of total mass three grams were prepared. For example, 1.86 g of surfactant was first weighed into a Pyrex tube vial (13x100 mm), then 0.09 g of deionized water was added and finally 1.05 g of Vitamin E (Fluka Chemical, # 355472/1 11098). Other samples made include 1.23 g of surfactant, 0.75 g of deionized water and 1.02 g of Vitamin E; and 0.69 g of surfactant, 2.16 g of deionized water and 0.15 g of Vitamin E. The sample tube vials were labeled and spun on the rotary wheel for ten minutes, and all formed clear microemulsions.

Microemulsions were also formed when Vitamin E was replaced with other polar oils. The successful compositions included 2.25 g of surfactant, 0.3 g deionized water and 0.45 g of Isopropylpalmitate (Lot # 97D31) and 1.5 g of surfactant, 0.9 g deionized water and 0.6 g of 1-octanol (Analysis # 366507/1 51697).

5

% DC5329/1,2-hexanediol	% H ₂ O	% Vitamin E	α	γ	Appearance
62	3	35	0.92	0.62	Clear
41	25	34	0.58	0.41	Clear
23	72	5	0.065	0.23	Clear

% Surfactant	% H ₂ O	% Polar Oil	α	γ	Appearance
75% 5329/1,2-hexanediol	10%	15% Isopropylpalmitate	0.6	0.75	Clear
50% 5329/1,2-hexanediol	30%	20% 1-Octanol	0.4	0.5	Clear

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Example IX

Dilution to form vesicles

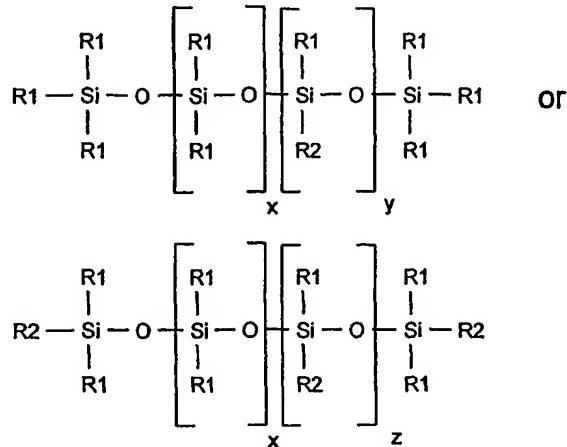
A microemulsion formed by method described in example VIII with 25% of 50:50 DC-5329 and 1,2-hexanediol, 70% H₂O and 5% vitamin E, was further diluted in at 1:3 dilution ratio. Figure 3 is the transmission electron micrograph (TEM) image of the formed vesicles.

A microemulsion composed of 35% 2-5329/1,2-Hexanediol (50/50), 40% H₂O, and 10% Vitamin was prepared by the method above. It was further diluted (1:7 dilution) in H₂O. TEM image showed vesicle formation, as shown in figure 4.

Claims

1. A composition comprising;

5 A) 0.01 to 90 wt % of a silicone surfactant having a formula



where R1 represents an alkyl group containing 1-6 carbon atoms;

R2 represents the group $-(\text{CH}_2)_a(\text{C}_2\text{H}_4\text{O})_b(\text{C}_3\text{H}_6\text{O})_c\text{R3}$;

x is 1-1,000; y is 1-500; z is 1-500; a is 3-6; b is 4-20; c is 0-5;

10 and R3 is hydrogen, a methyl group, or an acyl group;

B) 0.01 to 70 wt % of a co-surfactant selected from the group of a monohydroxy alcohol, an organic diol, an organic triol, an organic tetraol, a silicone diol, a silicone triol, a silicone tetraol, an cationic organic surfactant, an anionic organic surfactant, and an nonionic organic surfactant;

C) 0.01 to 60 wt % of a polar organic oil

20 D) and water, in an amount to provide the balance of the composition to 100 % on a weight basis,

wherein the weight ratio of components (A) to (B) ranges from 30:70 to 70:30.

2. The composition of claim 1 wherein the composition is a vesicle composition having a lamellar phase liquid crystalline structure comprising;

0.01 to 40 wt % of the silicone surfactant A),

0.01 to 60 wt % of the co-surfactant B),

5 0.01 to 40 wt % of the polar organic oil C),

and water in an amount to provide the balance of the composition to 100 % on a weight basis.

3. The composition of claim 1 wherein the composition is a microemulsion comprising;

10 7 to 90 wt % of the silicone surfactant A),

0.01 to 70 wt % of the co-surfactant B),

0.01 to 60 wt % of the polar organic oil C),

and water in an amount to provide the balance of the composition to 100 % on a weight basis.

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4. The composition of claims 1, 2, or 3 wherein the co-surfactant is an organic diol.

5. The composition of claim 4 wherein the co-surfactant is 1,2 hexanediol.

20 6. The composition of claims 1, 2, or 3 wherein the polar oil has an interfacial tension less than 40 mN/m vs water.

7. The composition of claim 6 wherein the polar oil is vitamin E or a vitamin E derivative.

25 8. A method for forming vesicles containing a silicone surfactant, co-surfactant, polar organic oil, and water comprising;

I) forming the microemulsion according to claim 3, and then

II) diluting the microemulsion with additional water to form vesicles.

30 9. A personal care formulation comprising any one of the compositions of claims 1-7.

5329/1,2-hexanediol (50/50)

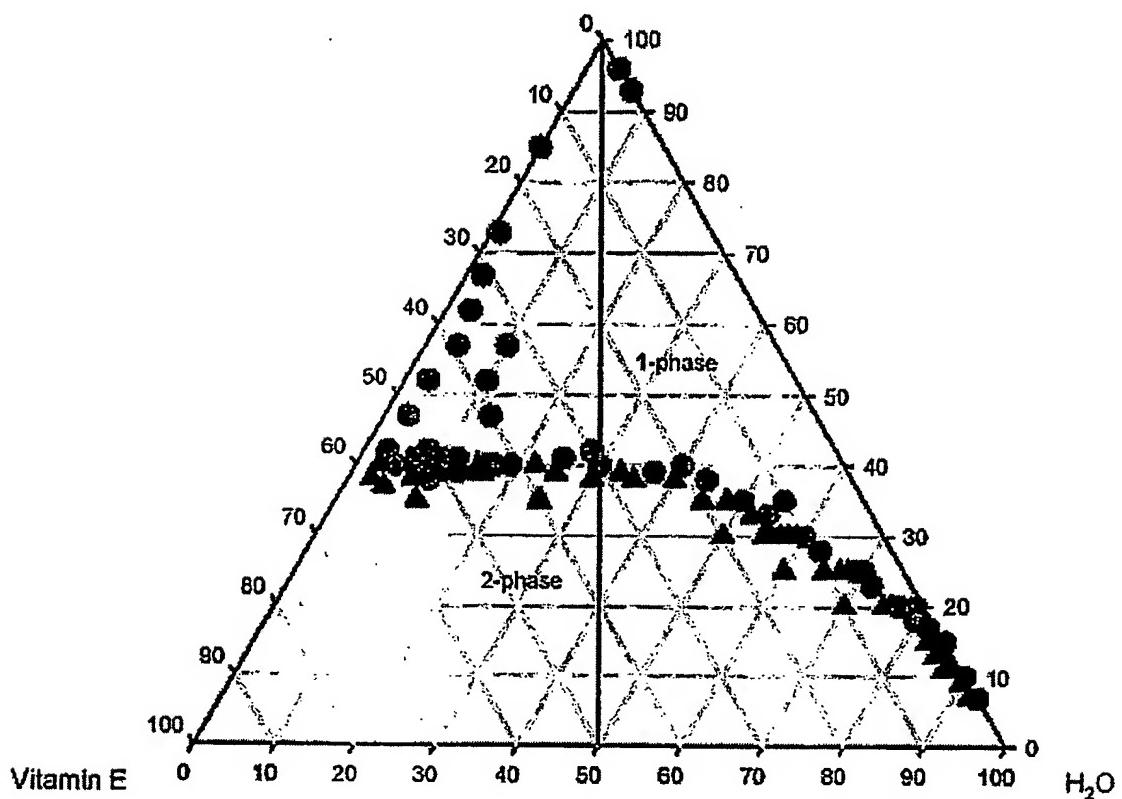


figure 1

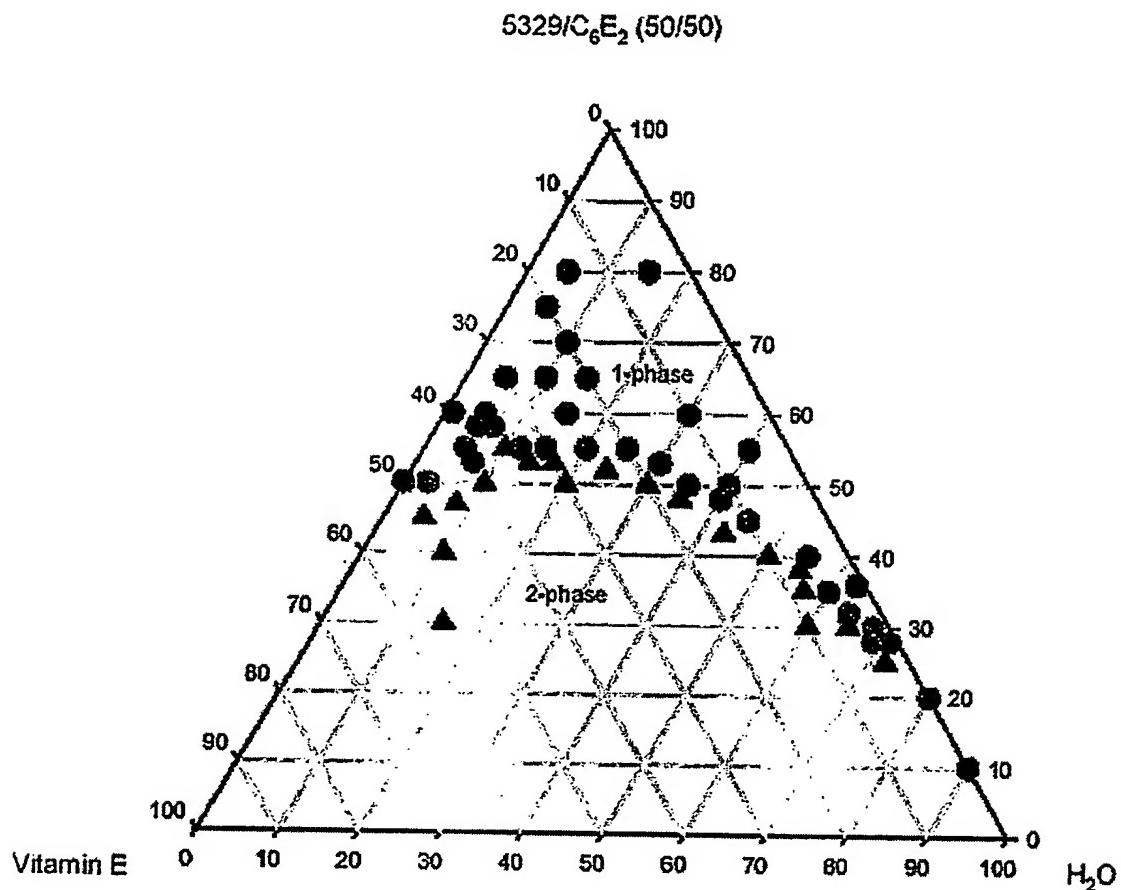
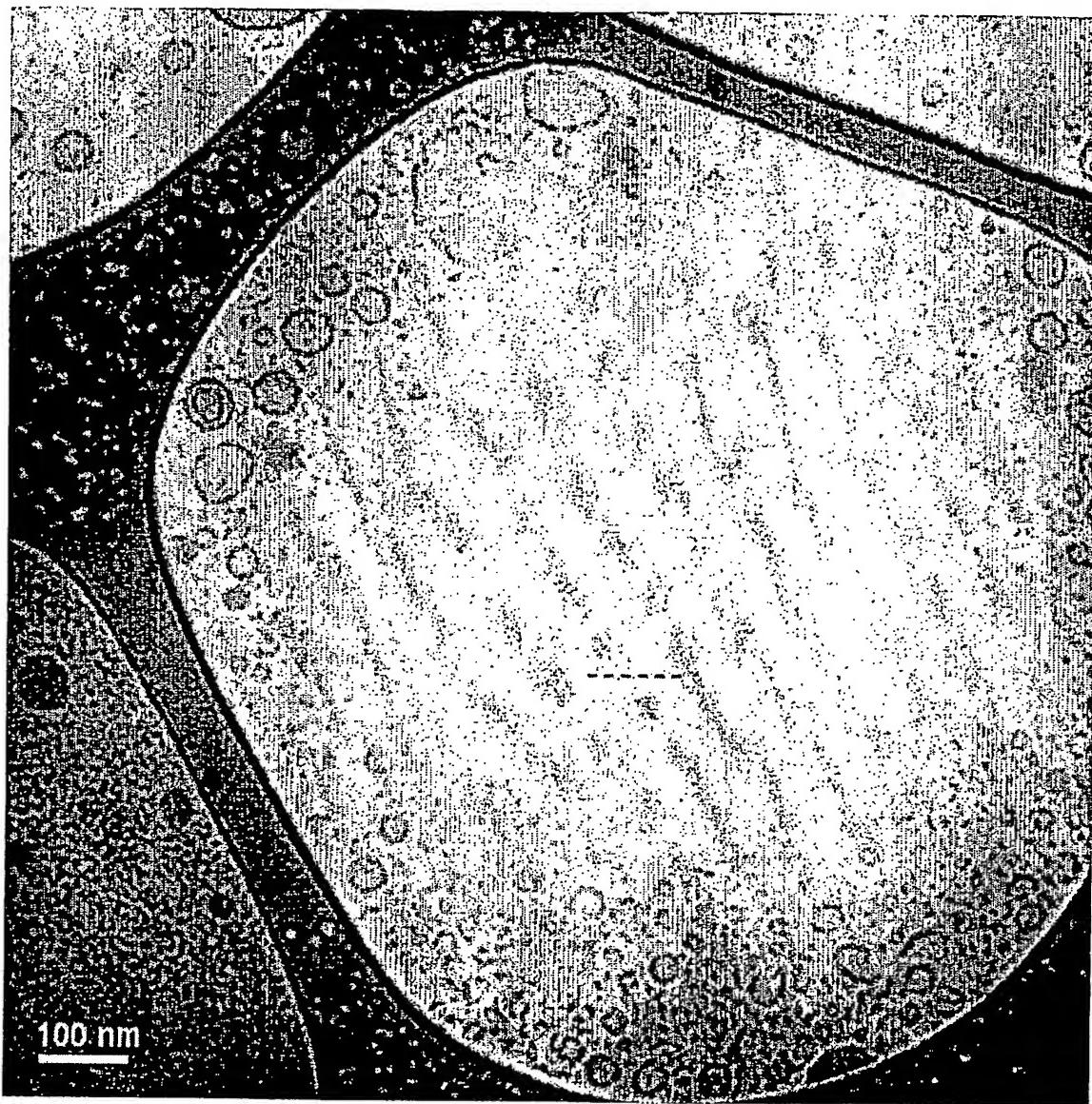


figure 2



5

figure 3



figure 4

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 03/38455

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 A61K7/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 299 112 A (BAILEY DONALD L) 17 January 1967 (1967-01-17) example 4 ---	1-7, 9
X	US 5 919 487 A (SIMONNET JEAN-THIERRY ET AL) 6 July 1999 (1999-07-06) cited in the application column 2, line 39-63; examples 1-5 ---	1-7, 9
X	WO 97/30690 A (UNILEVER PLC ;UNILEVER NV (NL)) 28 August 1997 (1997-08-28) example 5 ---	1-7, 9
X	EP 0 313 304 A (PROCTER & GAMBLE) 26 April 1989 (1989-04-26) example 9 ---	1-7, 9 -/-

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

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Date of the actual completion of the International search

19 April 2004

Date of mailing of the International search report

29/04/2004

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 03/38455

C(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 91/07943 A (GILLETTE CO) 13 June 1991 (1991-06-13) page 5, line 9-22; claim 1; examples 2,3 ----	1-7, 9
X	US 6 120 778 A (SIMONNET JEAN-THIERRY) 19 September 2000 (2000-09-19) cited in the application column 1, line 62 -column 3, line 37; claim 1; examples 1-3 column 4, line 27-57 column 5, line 46-49 ----	1-7, 9
Y	US 5 411 744 A (HILL RANDAL M ET AL) 2 May 1995 (1995-05-02) cited in the application column 1, line 14-18 column 3, line 7,8,28-45,55-61 column 5, line 64 -column 6, line 6 column 6, line 57-68 column 7, line 7 -column 8, line 44; claim 7 ----	1-9
Y	R.G. ALANY ET AL.: "Effects of alcohols and diols on the phase behaviour of quaternary systems" INTERNATIONAL JOURNAL OF PHARMACEUTICS, no. 196, 2000, pages 141-145, XP002276790 page 141 -page 143 page 145 -----	1-9

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
US 3299112	A	17-01-1967	BE DE FR GB NL NL US	665645 A 1493380 A1 1447461 A 1121831 A 133334 C 6507928 A 3359212 A	18-10-1965 30-01-1969 29-07-1966 31-07-1968 20-12-1965 19-12-1967
US 5919487	A	06-07-1999	FR DE DE EP ES JP JP	2742677 A1 69600758 D1 69600758 T2 0780115 A1 2125703 T3 3001821 B2 9175931 A	27-06-1997 12-11-1998 04-03-1999 25-06-1997 01-03-1999 24-01-2000 08-07-1997
WO 9730690	A	28-08-1997	AU WO	1596497 A 9730690 A1	10-09-1997 28-08-1997
EP 0313304	A	26-04-1989	US AT AU CA DE DE EP JP JP KR NZ	4847072 A 84703 T 2408388 A 1328409 C 3877690 D1 3877690 T2 0313304 A2 1265014 A 2812690 B2 131461 B1 226666 A	11-07-1989 15-02-1993 27-04-1989 12-04-1994 04-03-1993 27-05-1993 26-04-1989 23-10-1989 22-10-1998 17-04-1998 26-07-1991
WO 9107943	A	13-06-1991	AU DE DE EP ES WO	7172091 A 69026831 D1 69026831 T2 0503004 A1 2086531 T3 9107943 A1	26-06-1991 05-06-1996 12-12-1996 16-09-1992 01-07-1996 13-06-1991
US 6120778	A	19-09-2000	FR AT BR CN DE DE EP ES JP JP PL	2742676 A1 229792 T 9604724 A 1156586 A 69625452 D1 69625452 T2 0780114 A1 2188734 T3 3040355 B2 9175933 A 317635 A1	27-06-1997 15-01-2003 01-09-1998 13-08-1997 30-01-2003 12-02-2004 25-06-1997 01-07-2003 15-05-2000 08-07-1997 23-06-1997
US 5411744	A	02-05-1995	US DE DE EP JP	5364633 A 69509574 D1 69509574 T2 0672410 A1 7323222 A	15-11-1994 17-06-1999 07-10-1999 20-09-1995 12-12-1995

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